Comparative techno-economic modelling of large-scale thermochemical biohydrogen production technologies to fuel public buses: A case study of West Midlands region of England

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PII: S0960-1481(22)00222-1
DOI: https://doi.org/10.1016/j.renene.2022.02.074
Reference: RENE 16648

To appear in: Renewable Energy

Received Date: 21 July 2021
Revised Date: 2 February 2022
Accepted Date: 17 February 2022


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Danielle J. Nouwe Edou: contributed to the process simulation, process modelling, generation of economic analysis data and drafting of report and editing of the manuscript.

Jude A. Onwudili: contribution included project conceptualisation, methodology, synthesis and supervision of process design and economic analysis; project administration, and writing of the paper (draft, review and final editing).
Comparative techno-economic modelling of large-scale thermochemical biohydrogen production technologies to fuel public buses: A case study of West Midlands region of England

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Abstract

This work presents techno-economic modelling of four thermochemical technologies that could produce over 22,000 tonnes/year of hydrogen from biomass for >2000 public transport buses. These included fluidised bed (FB) gasification, fast pyrolysis-FB gasification, fast pyrolysis-steam reforming, and steam reforming of biogas from anaerobic digestion (AD). Each plant was modelled on ASPEN plus with and without carbon capture and storage (CCS), and their process flow diagrams, mass and energy balances used for economic modelling. Payback periods ranged from 5.10 to 7.18 years. For operations with CCS, in which the captured CO₂ was sold, FB gasification gave the lowest minimum hydrogen selling price of $3.40/kg. This was followed by AD-biogas reforming ($4.20/kg), while pyrolysis-gasification and pyrolysis-reforming giving $4.83/kg and $7.30/kg, respectively. Hydrogen selling prices were sensitive to raw material costs and internal rates of return, while revenue from selling CO₂ was very important to make biohydrogen production cost competitive. FB gasification and AD-biogas reforming with CCS could deliver hydrogen at less than or around $4/kg when CO₂ was sold at above $75/tonne. This study showed that four thermochemical technologies could contribute
to cheap biohydrogen production to extend the current use of electrolytic hydrogen-fuelled
buses in Birmingham to the wider West Midlands region.

Keywords: green biohydrogen fuel, FB gasification, pyrolysis-reforming, pyrolysis-gasification,
AD-biogas reforming, techno-economic modelling

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**Nomenclature**

- AD: Anaerobic Digestion
- ASPEN: Advanced Simulator for Process Engineering (Computer Software)
- BFB: Bubbling Fluidised Bed
- AD-BSR: Anaerobic Digestion-Biogas Steam Reforming
- CCS: Carbon Capture and Sequestration/Storage
- CFB: Circulating Fluidised Bed
- FB: Fluidised Bed
- FCI: Fixed Capital Investment
- H$_2$: Hydrogen
- HT: High Temperature
- IRR: Internal Rate of Return
- LT: Low Temperature
- Mtoe: Millions of Tonnes of Oil Equivalent
- NCF: Net Cash Flow
- NPV: Net Present Value
- PFD: Process Flow Diagram
- PSA: Pressure Swing Adsorption
- RME: Rapeseed Methyl Ester
- SMR: Steam Methane Reforming
- WGS: Water Gas Shift
1. Introduction

Increasing global population and improving living standards are major contributors to the exponential increase in global energy demand of approximately 14,000 Mtoe [1, 2] in 2019. Climate change and poor air quality are two of the most severe deleterious effects of our overwhelming dependence and utilisation of fossil fuels. Deployment of commercial-scale sustainable and renewable alternative fuels will help to meet the global 2050 decarbonisation and Net Zero targets [3] and tackle poor air quality in urban centres. Some urban areas, including Birmingham City, in the United Kingdom (UK) are using the Clean Air Zone scheme to discourage the use of old highly polluting vehicles within classified zones [4].

With no harmful emissions at the point of use and a potentially large availability of resources, hydrogen is now regarded as a promising ecologically clean energy carrier capable of ameliorating the long-term energy trilemma challenge. According to the EC Clean Vehicles Directive (Directive (EU) 2019/1161 by 2025 and 2030, EU countries must procure clean buses, including hydrogen-fuelled buses to meet binding emission targets) [5]. This has prompted several public and private initiatives to accelerate the pace of penetration of hydrogen-fuelled buses in Europe’s cities and regions. These include the H2BusEurope, JIVE, JIVE2, MERHLIN and 3MOTION schemes and projects that involve deploying about 1322 hydrogen-fuelled and fuel cell electric buses (FCEBs) across 22 cities across Europe by the early 2020s [6, 7].

Following Brexit, the UK is implementing subsisting or similar regulations that are well aligned to the EU Directives using schemes such as the FutureGrid and Project Union projects [8]. Hydrogen-fuelled public bus transport is being advocated in the UK, especially for large cities, including in Birmingham City [9]. By extension, these efforts should lead to replacing all public buses within the wider West Midlands region that are currently running on fossil fuels
(mainly diesel). With such initiatives, the demand for hydrogen energy is expected to increase dramatically, with an estimated global consumption anticipated to reach 300 million tonnes by 2050 [10,11].

Thus, producing hydrogen from an array of renewable sources is currently a hot topic. Depending on the feedstock and/or source of energy input for its production, the final hydrogen product can be colour-coded using a so-called hydrogen colour spectrum from black to white [12]. Generally, the commercial technologies for producing dark-coloured hydrogen (grey and black) include steam methane reforming (SMR) [13], partial oxidation (POX) of methane and other hydrocarbons [13] and coal gasification; all without carbon capture [14]. However, with a tail-pipe carbon capture and storage (CCS) and carbon capture, usage, and storage (CCUS), the colour of hydrogen product from these processes may move to lighter colour shades e.g., blue hydrogen from SMR with CCS. The ultimate green hydrogen comes from electrolysis of water powered with zero carbon emissions e.g., using wind or solar energy [15].

Biomass is a good feedstock for hydrogen production via different processing routes. Being renewable, biomass can become a source of green hydrogen, if the biomass feedstock is sustainably sourced and if the hydrogen production process is combined with CCS and CCUS [16]. Promising technologies for hydrogen production from biomass include conventional gasification [17,18], pyrolysis-gasification [19,20], pyrolysis-reforming [21,22], anaerobic digestion (AD) biogas reforming [23,24] and hydrothermal gasification [25–27]. Recent detailed reviews of these various technologies can be found in literature [18,28]. Within each technology space, different configurations and variants exist; for example, conventional gasification alone has several variants mainly depending on the type of gasifier and the gasifying agent [29]. For instance, in terms of gasifier type, conventional gasification can be classified into downdraft fixed bed, updraft fixed bed, entrained flow, circulating fluidised bed, bubbling fluidised bed and
plasma gasification [29,30]. However, the common feature of these technologies is the application of heat and various chemistries to convert biomass feedstocks into final products composed mainly of hydrogen and carbon dioxide, along with small amounts methane, carbon monoxide and even smaller amounts of other hydrocarbon gases [30,31].

In this present study, four different technologies for producing hydrogen from biomass have been selected for techno-economic evaluation based on established or growing commercial interests in their large-scale deployment [32–35]. Therefore, these four technologies arguably have the potential to meet the fuel requirement to power the current over 2000 buses being used for public transport in the West Midlands, UK [36,37]. Literature shows that previous researchers have modelled the techno-economic assessment of various hydrogen production technologies [38,39] however, few have combined all the viable biohydrogen production technologies within the context of a realistic public transport scenario. A combination of ASPEN modelling and techno-economic factors have been used to generate relevant process data in this study. It is the aim of this novel study to provide essential chemical engineering data for potential technology providers on large-scale biohydrogen production that will extend beyond the West Midlands public transport system towards meeting Net Zero targets.

2. Methodology

2.1. Design basis and biohydrogen requirement

In this section, a detailed outline of assumptions made for ASPEN simulation of the four biohydrogen production technologies (gasification, pyrolysis-gasification, pyrolysis-reforming and AD-biogas steam reforming), are presented, with and without CCS. The ASPEN Software used was ASPEN Plus V11 available at Aston University, Birmingham (UK). Appropriate feedstocks were selected for the different technologies based on literature and their availability
to meet the demand. Initial chemical process synthesis was carried out, followed by computation of process flowsheets to obtain mass and energy balances using ASPEN. Furthermore, the data obtained from the ASPEN simulation, along with conventional factors were used to evaluate the economic feasibility of using biohydrogen to meet West Midlands’ bus clean transport fuel demand. The impact of implementing CCS on the economic performance of each technology was also evaluated.

At present, the West Midlands currently operates over 2,000 buses, travelling a total distance of 515,000 km per day and consuming 230,000 litres of diesel [37,40]. Direct communication with Transport for West Midlands and Van Hool (one of the largest bus operators in Birmingham City), gave the number of public buses as 2000 and 2300, respectively [36,37]. Additionally, on average, 12-m buses consume 9 kg of hydrogen per 100 km [36]. Using these data, the yearly capacity of a hydrogen production plant operating for 330 days a year to produce fuel for 2000 buses was calculated as shown in Table 1. In addition, to account for the uncertainty in the actual number of buses and also accommodate potential future expansion, hydrogen production capacity was increased by a factor of 1.25, giving an adjusted hydrogen production rate of nearly 2782 kg/h.

Table 1: Case study data used for hydrogen production for 2000 buses [36,39, 40]

<table>
<thead>
<tr>
<th>Production Details</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average hydrogen consumption by buses</td>
<td>0.09</td>
<td>kg/km</td>
</tr>
<tr>
<td>Distance travelled by Bus</td>
<td>257.5</td>
<td>km/day</td>
</tr>
<tr>
<td>No. of Days the Bus is Driven</td>
<td>330</td>
<td>days/year</td>
</tr>
<tr>
<td>No. of Buses in West Midlands, England</td>
<td>2300</td>
<td>-</td>
</tr>
<tr>
<td>Yearly hydrogen requirement</td>
<td>17589.83</td>
<td>tonnes/year</td>
</tr>
<tr>
<td>Adjusted yearly hydrogen requirement</td>
<td>22031.78</td>
<td>tonnes/day</td>
</tr>
<tr>
<td>Yearly hydrogen plant operating hours</td>
<td>7920</td>
<td>hours/year</td>
</tr>
<tr>
<td>Adjusted hydrogen production capacity</td>
<td>2781.79</td>
<td>kg/h</td>
</tr>
<tr>
<td>Density of hydrogen at SATP</td>
<td>0.0813</td>
<td>kg/m³</td>
</tr>
<tr>
<td>Volumetric production capacity</td>
<td>34216.36</td>
<td>m³/h</td>
</tr>
</tbody>
</table>
2.2. Justification of selected process technologies

In this present study, thermochemical processing technologies for biohydrogen production, have been mostly considered due to their technical maturity, potential for large-scale application, high throughput capacity, fast reaction rates and robustness to meet the hydrogen requirement of 22,031.78 tonnes/year. All the processes considered are either at commercial scale or have the potential to reach commercial scale in the next few years. These include FB biomass gasification using circulating fluidised bed (CFB) reactor, combination of CFB pyrolysis and gasification (pyrolysis-gasification), combination of CFB pyrolysis and steam reforming (pyrolysis-reforming) and steam reforming of anaerobic digestion biogas (AD-biogas reforming).

Circulating fluidised beds are the most commonly used reactors in commercial gasification plants due to advantages of attaining isothermal conditions devoid of hotspots, excellent mass transfer rates and fast reaction rates [41]. Tar formation is a key challenge, but the use of dolomite and other catalysts can be used to induce tar cracking [42]. Furthermore, the process produces a syngas that mainly consists of hydrogen and carbon monoxide, with hydrogen making up around 40% of the volumetric fraction [39]. The relatively high CO composition of the product gas also gives opportunity for more hydrogen production via water-gas shift (WGS) reactions.

During pyrolysis-gasification biomass feedstock first undergoes fast pyrolysis upstream at 500 °C using a circulating bed pyrolyser, to produce up to 88 wt.% of vapour-phase products comprising of condensable and non-condensable compounds [43]. The vapour-phase products may be sent directly into a steam gasifier, to produce syngas containing hydrogen and for further downstream hydrogen production via a WGS reactor, condenser, scrubber, and a PSA unit [44]. Otherwise, the vapour-phase product may be separated by quenching and only the
liquid product (about 75 wt%) [43] re-vapourised and fed into the steam gasifier. Pyrolysis-reforming is another process route that upgrades the pyrolysis products using catalytic steam reforming. In most cases, cheap and readily available catalysts, such as Ni/Al₂O₃, are used to increase gas yields and thus, a higher production of hydrogen in the syngas from the pyrolysis vapours, largely based on SMR technology. Finally, anaerobic digestion (AD) is used to produce biogas, constituting of about 60% CH₄ and 40% CO₂ [45]. Once biogas is obtained, the CO₂ is removed and the methane-rich biogas is fed into a steam reformer for steam methane reforming (SMR), which is the current most widely used hydrogen production technology, generating around 50% of global hydrogen demand [46].

**Delimitations:** The evaluation of each model started with the feeding of solid biomass or biogas of the stated specification into the first thermal conversion equipment, respectively. Each model ended with: (1) the output of the required amount of hydrogen calculated from the design basis (2) where applicable, the combustion of char product to provide fractional process heats for the respective biomass conversion equipment (gasifier and pyrolyzer).

### 2.3. Feedstock selection and justification

Gasification, pyrolysis-gasification, and pyrolysis-reforming use biomass feedstocks in the form of wood chips and wood pellets, of which wood pellets are the most used. The UK produced only 0.3 million tonnes of wood pellets in 2020 and imported 9.1 million tonnes during the same period [47]. Therefore, the process plants based on direct thermochemical processing of biomass for biohydrogen production would rely on biomass imports. In contrast, AD could rely on abundant food and agricultural wastes generated within the UK, which generates around 13.1 million tonnes of food waste annually [48]. Therefore, UK has a relatively vast resource to
attain resource sufficiency for AD and in this study maize silage has been selected for the AD-biogas reforming plant.

2.4 Biomass feedstock characteristics

The average compositions of wood pellets (Supplementary Information A Table SI1) were obtained from literature [39,49] and used for the simulation of feedstock for hypothetical gasification, pyrolysis-gasification, and pyrolysis-reforming plants. Also, the ultimate compositions [50] of the maize silage used as feedstock for the AD-biogas process (Supplementary Information A Table SI1) but this was not used for simulation due to limitation of ASPEN Plus to simulate AD process. However, for simulation, the biogas produced from corn silage was used, since the biomass would be fed into an AD plant, which could not be simulated on ASPEN. Both feedstocks were assumed to have negligible sulphur contents based on literature data of around 0.1%.

2.5 Process description and syntheses

The simulation of each process flow diagram has been presented in Figures 1 - 4, with each having the additional optional CCS stage. Early trials indicated that the three thermochemical processes and the SMR part of the AD-biogas reforming process could be simulated on ASPEN Plus. Therefore, detailed process syntheses were carried out for the construction of process flowsheets with respect to the pieces of equipment to achieve the technical requirements for each process. The simulation of the AD process itself was considered beyond the scope of this present study, but details of biomass conversion and biogas yields for typical AD plants were obtained from literature [39] and used as appropriate.
2.5.1 FB gasification process

Physical properties for the non-conventional, conventional and CISOLID substances were calculated using IDEAL property methods. Furthermore, the enthalpy and density of the non-conventional components, (biomass, char, and ash) were all identified by the HCOALGEN and DCOALIGT models. The process flow diagram for the gasification process is presented in Figure 1. In the figure, an RYield block is used to portray the gasifier, operating at 850°C, where biomass reacts with steam at a steam to carbon molar ratio of 0.4 [39]. Thereafter, an SSplit block representing the cyclone separates the gaseous products (DEC-GAS) being transferred to the heat exchanger (HE), from the solid residues (DEC-CHAR) entering the RYield decomposition (DECOMP) unit. The DECOMP promotes the breakdown of biochar into C, N and ash which passes into an RStoic block where they react with air and the PSA recycle stream to undergo combustion to meet gasification energy demands. Therefore, at steady state the gasifier is assumed to be energetically self-sustaining. The oxygen to carbon molar ratio for the combustor is set at 0.3 [39] and maintaining the oxygen to nitrogen ratio of 21:79 in the feed air. A high temperature water-gas shift (HWGS) reactor was represented by an REquil block, while an Ryield block represented the low temperature water-gas shift (LWGS) reactor, and all heat exchangers by the heater block (Dryer, HE, HE1). Also, the RME scrubber was replicated by the Sep2 block. After separation, the hydrogen and carbon dioxide were compressed in the MCompr blocks (C-1, C-2). Heat transfer between the gasification units was also included within the simulation by heat streams (Q-SUPPLY, Q-SPARED).
Figure 1. An ASPEN Plus simulation depicting the Process Flow Diagram of Hydrogen Production from Biomass Gasification with Carbon Capture and Sequestration.

WBIOMASS=Wet Biomass; SEP= Separator; DBIOMASS= Dry Biomass; Q = Heat; DEC-GAS= Decomposed Gas; DEC-CHAR= Decomposed Char; DECOMP= Decomposition Reactor; COMBUST= Combustion Reactor; HWGS= High Temperature Water Gas Shift Reactor; LWGS= Low Temperature Water Gas Shift Reactor; CONWATER= Condensed Water; C-1= C-2= Compressor
2.5.2 Pyrolysis-gasification process

The physical properties, property methods and models were identical to that of gasification.

Figure 2 presents the process flow diagram for the pyrolysis-gasification process. The biomass was dried in a heater block (Dryer) at 110 °C to a moisture content of 7%. This was followed by an RYield fluidised bed fast pyrolysis reactor operating at 500 °C [49] under inert conditions, forming char, bio-oil, and gases. The products were assumed to be separated and only the bio-oil used for the gasification step; to eliminate the large volume fluidising gas (nitrogen) and process water. The normalised yields of the syngas, oil, char, ash and water products based on the work of Ringer et al. [49] were used for this stage. Additionally, inlet and outlet heat streams, $Q_{SUPPLY}$ and $Q_{SPARED}$, respectively, were inserted. The solid char was isolated using an SSplit cyclone unit and transferred to an RYield decomposition and RStoic combustor reactor at 600°C in series. Air was fed into the combustor at an air to carbon molar ratio of 1.15 [49]. A heater block at 850°C was used to heat up the captured bio-oil, prior to it being fed into the RYield fluidised bed gasification reactor. The steam reacted with bio-oil in the gasifier at a steam to carbon molar ratio of 2.75 [49]. The rest of the process, after the gasification stage, was assumed to be the same as described in biomass gasification.
Figure 2. An ASPEN Plus simulation highlighting the Process Flow Diagram of Hydrogen Production from Pyrolysis-Gasification with Carbon Capture and Sequestration.

WBIOMASS=Wet Biomass; SEP= Separator; DBIOMASS= Dry Biomass; HBIOMASS= Hot Biomass; PYROLYSI= Pyrolysis Reactor; Q = Heat Supply; DECOMP= DECOM1= Decomposition Reactor; COMBUST= Combustion Reactor; HBIO-OIL= Hot Bio-oil; DEC-CHAR= Decomposed Char; HWGS= High Temperature Water Gas Shift Reactor; LWGS= Low Temperature Water Gas Shift Reactor; CONWATER= Condensed Water; CO22= CO2, C-1= C-2= Compressor
2.5.3 Pyrolysis-reforming process

The starting point of this process was identical to the pyrolysis-gasification process, beginning with a fluidised based fast pyrolysis process as shown in Figure 3. Hence, a similar set of equipment was needed for this stage. After the pyrolysis stage, the reforming of the re-heated bio-oil would take place in an RGibbs catalytic steam reformer operating at 900 °C. The bio-oil would react with steam, at a steam to carbon molar ratio of 2.75 [49]. Gaseous products leaving the reformer would be cooled by heater block (HE2) to 450 °C, before entering the REquil HWGS reactor at 1.1 bar, followed by an LWGS at 250 °C. A Sep2 (CONDENS) unit would be used to remove water and other condensable components, leaving a gas stream being fed into optional Sep2 (scrubber) to remove CO₂, with the remaining gases compressed to 15 bar and 25 °C by an MCompr block (C-1). The specification for the HWGS was the same as that for both biomass gasification and pyrolysis-gasification. The compressed gases would be fed into another Sep2 (PSA) block, where an assumed 99.9% of hydrogen gas would be obtained.
Figure 3. An ASPEN Plus simulation picturing the Process Flow Diagram of Hydrogen Production from Pyrolysis-Reforming with Carbon Capture and Sequestration.

WBIOMASS=Wet Biomass; SEP= Separator; DBIOMASS= Dry Biomass; HBIOMASS= Hot Biomass; PYROLYSI= Pyrolysis Reactor; Q-SUPPLY= Heat Supply; Q = Heat Spared; DECOMP = Decomposition Reactor; COMBUST= Combustion Reactor; HBIO-OIL= Hot Bio-oil; SMR= Steam Methane Reforming Reactor; HWGS= High Temperature Water Gas Shift Reactor; LWGS= Low Temperature Water Gas Shift Reactor; CONWATER= Condensed Water; C-1= C-2 = Compressor
2.5.4. AD-Biogas steam reforming process

As stated at the beginning of this section, the AD-biogas steam reforming process simulation would start with the biogas, as ASPEN could not be used to replicate the enzymes used in AD. The BSR process flow diagram is shown in Figure 4. Therefore, the starting point for simulation was the compression of biogas in an MCompr block (C-1). Yields of the products of anaerobic digestion found in literature [45, 50] were normalised before being entered into ASPEN (Supplementary Information A Table SI2).

It was assumed that biogas only consists of CH₄ and CO₂ so only the normalised values for these two components were used in the simulation (Supplementary Information A Table SI2). A Sep2 (scrubber) was used to isolate CO₂ whilst the remaining CH₄ was heated by a heater block (HE) and transferred to the RGibbs (SMR reactor) block at an operating temperature of 900 °C. The reaction of methane with steam would occur at a steam to carbon molar ratio of 3 [39]. Gases leaving RGibbs were cooled to 350 °C by a heater block (HE1) and again, following the same steps detailed under FB gasification. The Redlich-Kwong-Soave cubic equation with Boston-Mathias alpha function (RKS-BM) property method was used, with the density and enthalpy values calculated using the same models as the previous processes.
Figure 4. An ASPEN Plus simulation illustrating the Process Flow Diagram of Hydrogen Production from Biogas Steam Reforming with Carbon Capture and Sequestration.

CBIOGAS = Compressed Biogas; HE = Heat Exchanger; H-BIOGAS = Hot Biogas; SMR = Steam Methane Reforming Reactor; HWGS = High Temperature Water Gas Shift Reactor; LWGS = Low Temperature Water Gas Shift Reactor; CONDENS = Condenser; CONWATER = Condensed Water; C-1 = C-2 = C-3 = Compressor, PSA = Pressure Swing Adsorption
For all four processes, an optional unit of an MCompr block (C-2) at 55 bar and 25 °C used to compress and capture CO₂ was included. The flowsheets for processes without carbon capture and storage (CCS) can be found in the Supplementary Information B.

2.6. Economic analyses

This section summarises the description of the approach taken for the techno-economic evaluation of each process route. The following assumptions were used for cost estimation for the base case scenario:

- Depreciation was charged over 10 years [51].
- Income tax rate was estimated to be 30% [39,51].
- Ideal breakeven point was assumed to be 40% [52].
- Fluctuations in costs due to general inflation or processing demands were negligible when calculating the Net Present Value [52].
- Baseline case discount rate or required return was assumed to be 15% [53].
- Where applicable, CO₂ selling price ranged from $0 to $150 per tonne [54]
- Wood pellets price was taken as $150 per tonne and maize silage as $45 per tonne; both on ‘as received’ basis.

2.6.1. Capital cost

All purchased equipment costs were identified using a combination of the ASPEN Process Economic Analyser software and techno-economic studies from literature [52,53]. A detailed factorial method was used to estimate the capital cost of the biomass gasification using US Gulf Coast Basis [52]. Additionally, location factors for USA and the UK were applied to find the realistic capital cost of the same plant in the UK. For instance, published price data from
Chemical Engineering Economics [55] were used for fluidized/packed bed reactors to determine cost of equipment. To obtain prices for 2020 (the start of this study), the Oil and Gas Field Machinery, and Equipment Manufacturing Cost Index [56], published in 1989 was used according to Equation 1.

\[ C = C_i \times \frac{{\text{Index}_{2020}}}{{\text{Index}_{1989}}} \times f \]  

(1)

Where, \( C \) = present cost of equipment, \( C_i \) = historical cost of equipment, \( f \) = factor accounting for construction material type and insulation (\( f = 1.30 \) for stainless steel).

2.6.2. Operating costs

Operating costs in process plants are typically made up of ongoing costs including raw materials costs, maintenance costs, utility costs and labour costs. Apart from feedstock (biomass) costs, the other operating costs have been calculated using typical Lang factors [52] (Supplementary Information A Table SI3).

2.6.3. Profitability parameters

The key economic parameters were calculated [52] using the usual formulae (Supplementary Information A Table SI4). These include depreciation, net cash flow (NCF), breakeven point, payback period, net present value (NPV) and minimum hydrogen selling price. In this study, three main scenarios were considered for each of the four selected technologies: (a) plant operations with CCS and selling captured CO\(_2\); (b) operation with CCS but without selling the captured CO\(_2\); and (c) operation without CCS. The breakeven point was expected to occur at below 70% [32] of the operating capacity to still make profit while accommodating fluctuations in operational contingencies and market demands. Finally, for each scenario, the selling price of H\(_2\) was adjusted to obtain a net present value (NPV) of zero to obtain the minimum selling
price of hydrogen for comparison with other commercially viable hydrogen production technologies.

3. Results and Discussion

3.1 Mass balances

In the chemical industry, raw materials account for 80-90% of the total cash cost of production (CCOP) [57] and it is therefore an important factor to consider during economic evaluation of chemical processes. The biomass feedstock requirements based on ASPEN simulation for each technology are presented in Table 2 as part of Blackbox mass balances. The table shows that the process requiring the largest amount of raw material was gasification with a total of 93,784.84 kg/h biomass (wet basis) to produce the required 2,781.79 kg/h of hydrogen. On the other hand, the technology requiring the least biomass was pyrolysis-gasification, with a total initial biomass quantity of 29,013.67 kg/h (wet basis). The biomass requirement for AD of 69,905.30 kg/h (wet basis) was estimated on the basis of the quantity of biogas and typical conversion efficiencies needed to make the required amount of biomethane to produce the stated amount of hydrogen [39].

When combined in series with other upstream processes such as pyrolysis, gasification became more attractive as less raw material (a decrease of 69.1%) was required as seen in the case of pyrolysis-gasification. Moreover, combining pyrolysis in series with reforming required similar biomass feedstock as pyrolysis-gasification. The dramatic reduction in feedstock requirement compared to gasification could be attributed to the increased contribution of hydrogen atoms in the reactant steam to eventual hydrogen gas product [25].

The carbon cost of biohydrogen production via thermochemical technologies lies in the inevitable co-production of CO₂, which can influence their economic and environmental
performance [58]. CO₂ co-production thus has implications for environmental sustainability (if emitted) and economic viability (if sold) on biomass-based hydrogen production technologies. Clearly, since the gasification process consumed the most biomass H/C ratio = 1.5 in Table SI1), it meant that more of the carbon element would be present throughout. Thus, a greater mass flowrate of carbon dioxide was produced, totalling 705,473 tonnes/year. As expected, the CO₂ production rate decreased with a reduction in biomass feedstock requirement, hence the drop to 408,522 tonnes/year and then 316,801 tonnes/year for pyrolysis-gasification and pyrolysis-reforming, respectively. AD-biogas steam reforming gave a much lower direct CO₂ production rate per annum of 234,806 tonnes/year due to using mainly methane (H/C ratio = 4) for hydrogen production. The large participation of steam in hydrogen production from the reforming-based technologies, meant that they required lower quantities of biomass feedstocks and thus generating lower CO₂ co-product than gasification-based technologies. Char combustion in the three solid biomass thermal conversion technologies, will emit further CO₂ (Supplementary Information A Table SI5). As char combustion would only provide a fraction of the process heat required for each gasifier and reformer (Supplementary Information A Table SI6), additional biomass combustion would be needed provide the balance. Therefore, CO₂ emissions from the additional biomass combustion must be included in life-cycle analysis studies, which is beyond the scope of this present study. The detailed mass balance calculations and enthalpy flows (heat balances) for each technology, with or without CCS, are given in Supplementary Information B.
Table 2: Blackbox mass balances for all four hydrogen production technologies

<table>
<thead>
<tr>
<th>Streams</th>
<th>Stream Name</th>
<th>FB Gasification With CCS</th>
<th>Pyrolysis-Gasification With CCS</th>
<th>Pyrolysis-Reforming With CCS</th>
<th>Biogas Steam Reforming With CCS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inlet (kg/h)</td>
<td>Wet biomass</td>
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<td>30,504.24</td>
<td>(69,905.30)</td>
</tr>
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<td></td>
<td>Dry biomass</td>
<td>(71,049.12)*</td>
<td>(19,342.45)*</td>
<td>(20,336.16)*</td>
<td>(24,466.85)*</td>
</tr>
<tr>
<td></td>
<td>Steam</td>
<td>21,058.06</td>
<td>40,634.09</td>
<td>42,721.65</td>
<td>18,870.01</td>
</tr>
<tr>
<td></td>
<td>Biogas</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>18,232.68</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td>114,842.90</td>
<td>69,647.76</td>
<td>73,225.89</td>
<td>37,102.69</td>
</tr>
<tr>
<td>Outlet (kg/h)</td>
<td>Exhaust from biomass dryer</td>
<td>11,367.86</td>
<td>11,367.86</td>
<td>9,456.32</td>
<td>9,456.32</td>
</tr>
<tr>
<td></td>
<td>Char (+ Ash)</td>
<td>6006.84</td>
<td>6006.84</td>
<td>3,411.87</td>
<td>3,411.87</td>
</tr>
<tr>
<td></td>
<td>Condensed water</td>
<td>1,909.99</td>
<td>1,909.99</td>
<td>1,035.46</td>
<td>1,035.46</td>
</tr>
<tr>
<td></td>
<td>H₂ Product</td>
<td>2,781.79</td>
<td>2,781.79</td>
<td>2,781.79</td>
<td>2,781.79</td>
</tr>
<tr>
<td></td>
<td>Recycle gases</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>CO₂ to Storage</td>
<td>89,074.89</td>
<td>40,000.07</td>
<td>51,581.09</td>
<td>29,647.17</td>
</tr>
<tr>
<td></td>
<td>Off-gases</td>
<td>3,701.54</td>
<td>92,776.43</td>
<td>51,659.09</td>
<td>112.59</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td>114,842.90</td>
<td>69,647.76</td>
<td>73,225.89</td>
<td>37,102.69</td>
</tr>
</tbody>
</table>

* Data in parenthesis have not been used in the mass balance calculations
3.2. Enthalpy flows (heat balances)

ASPEN Plus simulation (Supplementary Information A Table SI6) was used to calculate the enthalpy flows (heat balances) for the main biomass conversion equipment for each process as well as heat provided via residual char combustion, except for AD-biogas reforming. Even with char combustion in applicable technologies, all four technologies presented net positive heat balances due to the main initial biomass or biogas conversion reactions all being highly endothermic. Pyrolysis-gasification gave the highest net heat balance of +101,906.19 kW, due to the presence of both the pyrolyzer and gasifier, which require high heat inputs. Char combustion could be used to provide significant amount of heat energy for the main feedstock conversion reactors, except for AD-biogas reforming (Table SI6). Literature has shown that solid biomass thermal conversion technologies can be energetically self-sufficient by combusting some of the biomass and the inevitable char product [17]. From literature, the char combustor efficiency of 98% was assumed, with flue gas temperature of 1200 °C [59]. The heat exchange temperature approach for the gasifier was assumed to be 100 °C. Therefore, assuming the flue gas temperature reduced to 950 °C after heating the gasifier operating at 850 °C, the heat balance of the flue gas across the gasifier was calculated to be -19,368.83 kW, using Equation 2.

\[ \dot{Q}_{\text{flue gas}} = \sum_{i=1}^{n} n_i C_{p,i} (T_2 - T_1) \]  

\[ \dot{Q}_{\text{flue gas}} = \text{enthalpy balance of flue gas} \]

\[ n_i = \text{molar flow rate of flue gas component } i \]

\[ C_{p,i} = \text{average molal specific heat capacity of flue gas component } i \]

\[ T_2 = \text{flue gas temperature after heating gasifier} \]

\[ T_1 = \text{temperature eof flue gas xiting combustor} \]
Similar calculations were performed for the pyrolyzer (operating at 600 °C). As shown in Table SI6, the hot flue gas from the combustion of char could be used to offset some of the process heat, thereby reducing the overall process heat requirements. This will also reduce the amount of biomass combusted for process heat around the solid biomass conversion equipment (gasifier and pyrolyzer). In addition, the spent flue gas (still at 950 °C) can be used to pre-heat the combustion air and/or used in a waste heat boiler to raise steam, as part of energy integration. Detailed analysis of the energy efficiencies and influence on hydrogen selling price across each technology would include other forms of energy (mechanical and electrical), which is outside the scope of this present study. The use of typical Lang factors [52] to represent cost contributions from utilities and services (including energy) obviates the need for such complicated analysis in this type of study.

3.3 Economic modelling results

This section presents an economic evaluation of each of the four selected hydrogen production technologies by obtaining the minimum hydrogen selling price for each scenario studied. The key parameters used included the capital costs, operating costs, minimum hydrogen selling price, CO$_2$ selling price, cumulative net cash flow, net present values, breakeven points, and payback periods. Economic analyses were carried out with and without CCS for each process using a baseline case of 15% discount rate, which is typical of bioenergy projects [53]. In addition, sensitivity analysis results on the influence of raw material costs, discount rates and CO$_2$ selling price on the minimum selling price of hydrogen.

3.3.1 Total production costs
Through comparison between the costs provided by the computer software and data obtained in literature [60, 61] a final purchasing cost for a circulating fluidised bed (CFB) reactor was calculated by order of magnitude for gasification, pyrolysis-gasification and pyrolysis-reforming, respectively. An example of evaluated total capital expenditure for each selected hydrogen production technology (with CCS) can be found in the Supplementary Information A (Table SI7). The overall production costs and other economic parameters used for the modelling of the four plants under different scenarios were also calculated (Supplementary Information A Table SI8).

Figure 5 shows the total production costs required for each technology to produce the 22,032 tonnes per year of hydrogen needed to fuel all 2,000 buses in West Midlands, England. The TPC was broken down into capital and operating costs as shown in Figure 5. As mentioned in Section 2.5.4, the ASPEN simulation of the AD-biogas reforming technology did not include the upstream AD plant, however, the purchased cost of AD was obtained from literature [62] and included in the economic analysis. Even with the addition of this unit, AD-biogas reforming remained the cheapest process with a capital cost of $101.24 million with CCS. In a similar way, FB gasification with fewer processing equipment also gave a lower capital cost than pyrolysis-gasification and pyrolysis-reforming. Unsurprisingly, pyrolysis-gasification with and without CCS were found to be cheaper than pyrolysis-reforming by $54.69 million and $43.03 million, respectively.
Figure 5. Hydrogen production costs for all four technologies without and with CCS (a) capital costs; and (b) operating costs.

A similar trend was also observed for the operating costs, such that biogas-steam reforming gave the lowest operating costs, followed by FB gasification. Even though FB gasification required the largest quantity of biomass feedstock to produce the required amount of hydrogen (Table 2), it still gave a lower operating cost than pyrolysis-gasification and pyrolysis-reforming, respectively, indicating that the complexity of the processes contributed more to the operating costs. Therefore, at the scale required, the two hyphenated thermochemical biomass conversion technologies (pyrolysis-gasification and pyrolysis-reforming), requiring two large
initial reactors each, appear to be highly expensive compared to FB gasification and AD-biogas reforming.

Between the two hyphenated thermochemical processes, pyrolysis-reforming was much more expensive than pyrolysis-gasification, with or without CCS. With CCS, the capital cost and operating costs of pyrolysis-reforming were, respectively, 24.6% and 27.6% higher than pyrolysis-gasification. Without CCS, pyrolysis-reforming was also 24.6% and 23.3% higher than pyrolysis-gasification in terms of capital cost and operating costs, respectively. While Table 2 shows that both pyrolysis-gasification and pyrolysis-reforming required similar quantities of biomass feedstock and steam, the large difference in total production costs is due to the overall complexity of pyrolysis-reforming technology.

3.3.2 Minimum hydrogen selling price

The minimum hydrogen selling price was obtained when the NPV of each process became zero, indicating that above that selling price, NPV became positive, which was deemed sufficient to determine the feasibility of each technology at the early stage of engineering projects [52]. For the baseline case, CO₂ selling price was fixed at $120 per tonne (where applicable), which according to Salkuyeh, Saville and Maclean [63], should be the least selling price of CO₂ to make biohydrogen production from thermochemical technologies cost competitive with SMR (the cheapest hydrogen price in the market).

Figure 6 presents the minimum selling prices of hydrogen product for the selected technologies. Results have been evaluated with a consideration to sell or not to sell the captured CO₂ in operations with CCS. Selling CO₂ to obtain low hydrogen selling price may become useful in future with the advancement in technology for CO₂ utilisation, with potential large-scale CO₂ market in the horizon. The CO₂ could be sold to the brewing industry, algae cultivation industry
or used for fuel and chemical production; the latter being a subject of increasing research interest. In all cases, breakeven points occurred below 50% operating capacity of each plant (Supplementary Information A Figures SI1 and SI2), indicating huge capacities for the plants to make profit, if the minimum hydrogen selling prices could be competitive. In addition, the payback periods for each scenario for each plant were between 5.10 and 7.18 years (Supplementary Information A Figure SI3) However, lower payback periods were obtained with the implementation of CCS due to increased income from CO$_2$ sales, which supersedes the additional CCS investment. At the scale of production used in this study, pyrolysis-reforming, and without CCS and with CCS and CO$_2$ sold, gave lowest payback periods of 5.51 and 5.10 years, respectively. Typical payback periods for biomass conversion plants are 2 – 7 years [64,65]. Therefore, all the four technologies (with CCS) selected for this present study were within the typical range of typical payback periods.

With CCS and CO$_2$ being sold as a co-product, all four technologies gave hydrogen selling price of between the lowest value of $3.40/kg for FB gasification and the highest value of $7.30/kg for pyrolysis-reforming. Figure 6 however, shows the more present realistic scenarios of CCS without selling CO$_2$ (e.g., sequestration in old oil formations – with added transport cost not considered in this study) and without CCS at all. For both scenarios, CCS with no CO$_2$ sold gave slightly higher hydrogen selling price than without CCS, due to the additional total production costs involved in CCS. Capturing CO$_2$ without selling it still represented the better option to make biohydrogen production greener and comparable to other low-emission hydrogen production routes (e.g., electrolysis of water). Therefore, in this scenario, AD-biogas reforming gave the lowest minimum hydrogen selling price of $4.75/kg, followed by FB gasification at $5.50/kg, with pyrolysis-reforming and pyrolysis-gasification giving $8.10/kg and $6.1/kg, respectively. Although, Figure 6 shows that minimum hydrogen selling prices were
fractionally lower for all four technologies without CCS, this scenario should be deemed as being environmentally unsustainable and therefore not encouraged.

Figure 6. Hydrogen selling price for each production technology without and with CCS (* selling the captured CO$_2$; ** not selling the captured CO$_2$)

These results show that at scale, both FB gasification ($3.4/\text{kg}$) and AD-biogas reforming ($4.20/\text{kg}$) compared favourably with the current market prices of biohydrogen from manufacturers such as the Canadian company, H$_2$ V Energies, who sell hydrogen at $2.67/\text{kg}$ [66]. In addition, Salkuyeh, Saville, and MacLean [43] obtained a value of $3.10/\text{kg}$ as the minimum selling price of hydrogen from a fluidized bed gasification process (FB gasification), which is similar to the value obtained from this present study.

Although, the minimum selling prices from the four technologies were still higher than the selling price of highly polluting grey hydrogen from SMR at $1.50$ [67], significant future investments could help make them more competitive. In comparison, green hydrogen from water
electrolysis, currently sells at around $12/kg and future sustainable prices have been predicted to be between $6 and $7 per kg depending on source of electricity [67]. Therefore, this present study shows that thermochemical production of biohydrogen with CCS is potentially a relatively cheap route to green hydrogen using FB gasification and AD-biogas reforming, whereas green biohydrogen from pyrolysis-gasification and pyrolysis-reforming are at close parity to water electrolysis. However, the large quantities of CO$_2$ generated from these technologies must deliver economic value through sales in order for these biohydrogen routes to be highly successful economically. Hence, there is a wide range of research activities going on to find economic uses of CO$_2$.

3.4 Results of sensitivity analyses

3.4.1 Influence of raw material costs

Figure 7 shows the influence of varying the raw material costs on the minimum hydrogen selling price. The costs of the main raw materials (biomass feedstock, water, steam and catalysts) have been varied by +/-20% of the nominal values obtained from the initial economic modelling of the four hydrogen production technologies. The range have been chosen as realistic scenarios considering that with the limitations on economic production of biomass feedstocks, it is highly unlikely that prices would fluctuate outside this range by so much. The evaluation was based on the three scenarios used in this present study. With raw material costs falling by 20%, the minimum hydrogen selling prices also decreased compared to those obtained with the nominal raw materials costs. In the case of CCS with CO$_2$ sold, hydrogen minimum selling price fell by 7.6% for pyrolysis-reforming, 8.1% for pyrolysis-gasification, 8.2% for AD-biogas reforming and 12% for FB gasification. These changes in hydrogen minimum selling prices reflected the differences in the quantities of biomass required for each technology. Therefore,
with FB gasification requiring the most biomass, the effect of changing raw material costs would have the most impact on the minimum hydrogen selling price from this technology. Hence, the minimum hydrogen selling price fell to $2.50/kg for FB gasification, which although still higher than hydrogen from SMR, was nearly 5 times cheaper than the current price of hydrogen from water electrolysis ($12/kg) [68].

It would also remain cheaper even at the future green hydrogen price of between $6 and $7 per kg, provided CO₂ sales could be guaranteed. When the raw material costs were increased by 20%, a similar trend was observed where the minimum selling price of hydrogen again increased by 12% for FB gasification, 7.9% for pyrolysis-gasification, and 7.6% each for pyrolysis-reforming and AD-biogas reforming. For the other scenarios, a similar trend was observed with hydrogen minimum selling prices decreasing when raw materials costs fell by 20% and also increasing when they increased by 20%. These results show that raw material costs can have significant influence on hydrogen selling prices for all four technologies and through all the studies scenarios.
Figure 7. Influence of changes in raw materials cost on the minimum selling price of hydrogen from the different technologies (a) at -20%; and (b) at +20% of nominal values.

3.4.2 Influence of discount rate (internal rate of return)

The influence of internal rate of return (IRR) on the minimum hydrogen selling price is presented in Figure 8. In this study, the internal rate of return was used as discount rate, assumed as the expected rate of return and then used to find the minimum selling price of hydrogen that gave
a net present value of zero for each selected technology and scenarios. The IRR was used at 10%, 15% (baseline case) and 20% for this present study. Most bioenergy projects are considered high-risk due to lack of long years of technical experience and IRR values can vary widely, however, for waste-to-energy plants, IRR values of around 10% - 15% are seen as acceptable [53, 69,70].

It is clear from Figure 8 that the minimum hydrogen selling prices increased linearly for each technology and each scenario. In all three cases, FB gasification with CCS and CO₂ sold, gave minimum hydrogen selling prices of $2.52/kg at 10%, $2.84/kg at 15% (baseline case) and $3.2/kg at 20% IRR, respectively. Similarly, AD-biogas reforming with CCS and selling the captured CO₂ gave minimum hydrogen selling prices below $4/kg ($3.42/kg at 10%, $3.67/kg at 15% and $3.95/kg at 20%).

Figure 8. Influence of changes in internal rate of return on the minimum selling price of hydrogen from the different technologies (* with CCS, CO₂ sold; ** with CCS, CO₂ not sold; *** without CCS)
Therefore, for both technologies, investors could expect to be cost competitive with attractive IRR of up to 20%. While higher IRRs could be potentially obtained, keeping the minimum selling price of hydrogen around $3/kg or below is important for these technologies to be viable and competitive. For pyrolysis-gasification and pyrolysis-reforming, the minimum hydrogen selling prices remained above $6/kg which seemed high but could be competitive with green hydrogen from water electrolysis.

3.4.3 Influence of CO₂ selling price

As mentioned in Section 3.3, CO₂ production is an investable consequence of hydrogen production from any carbon-based feedstock, including biomass. So far, results have shown that the sale of the CO₂ co-product has significant impact on the minimum hydrogen selling price for each technology. Clearly, with CO₂ captured via CCS and sold of economic utilisation, hydrogen selling price could be lowered to make it more competitive with SMR. Although, CCS would add to the TPC of each technology, the revenue from selling the CO₂ can outweigh this added cost and deliver cheaper biohydrogen. Figure 9 presents the influence of varying the selling price of the captured CO₂ from $0 to $200 per tonne. The results in Figure 9 also demonstrate the effect of the quantity of biomass used and therefore CO₂ expected from each technology. For instance, FB gasification required the largest amount of biomass (93,784.84 kg/h in Table 2) and therefore would produce the largest quantity of CO₂ both from the process stream and from char combustion for heat energy. Indeed, Figure 9 shows that the minimum hydrogen selling price fell sharpest with increasing CO₂ selling price for FB gasification and this rate of fall was followed by pyrolysis-gasification, which required the second largest biomass feedstock on dry basis (Table 2).
Figure 9. Influence of CO$_2$ selling price on the minimum hydrogen selling price for the different technologies.

For the reforming technologies, the rate of decreasing minimum hydrogen selling prices were slower due to the large contributions of water in hydrogen production in these processing, which ultimately reduced their biomass feedstock requirements. For FB gasification, the hydrogen selling price fell below $4/kg when the CO$_2$ selling price was about $75/tonne, while for AD-biogas reforming, this could also be achieved around a CO$_2$ selling price of $80-85$ per tonne.

As seen earlier, these two technologies have the better chances of being cost competitive. However, for pyrolysis-gasification and pyrolysis-reforming, the minimum hydrogen price could only break below $6/kg with a CO$_2$ selling price of $160/tonne (pyrolysis-reforming) and $190/tonne for pyrolysis-gasification. Although these two hyphenated thermochemical technologies appeared to deliver higher minimum hydrogen selling prices, they could compete with hydrogen from water electrolysis. In addition, they both have the advantage of using much
lower biomass feedstock compared to FB gasification and may also have the technical advantage of reduced problems with ash and tar at the syngas production stage compared to FB gasification [71].

4. Conclusion

The slow introduction of hydrogen fuel has resulted from major barriers regarding high feedstock costs, lack of infrastructural development for its production and transport, which appear to outweigh its practical environmental and sustainability benefits. However, it has become apparent that with further supportive policies from governments into the research and development of different thermochemical biohydrogen production technologies, which have the best chances of large-scale capacities, they may attain commercial viability sooner than later.

In this study, to identify the most feasible of these technologies process, simulations with and without CCS followed by techno-economic assessments were undertaken. ASPEN Plus was used to simulate the following processes: FB gasification, pyrolysis-gasification, pyrolysis-reforming, and biogas steam reforming.

All four technologies are energy-intensive and, apart from AD-biogas reforming, some of the energy requirements can be satisfied from the combustion of the resultant char formed during the initial biomass conversion stage. Despite this fact, FB gasification and AD-biogas reforming could deliver hydrogen at minimum selling prices that are much lower than that currently obtained from water electrolysis. Results show that hydrogen selling prices were sensitive to CO₂ selling price, raw material costs and IRR. However, capturing and selling the CO₂ coproduct appeared to be the most important economic factor in diversifying the income streams in order to deliver hydrogen at affordable prices. Although, uncertainties remain in term of the influence of inflation on feedstock prices, equipment costs and labour costs, the scenario
presented in this work indicate that biohydrogen production from these technologies, especially FB gasification and AD-biogas reforming, can be delivered affordably and sustainably to power all the public transport buses in West Midlands, England. Technical challenges such as tar formation remain with FB gasification, which could be overcome via pyrolysis-reforming, but this technology require further research and development to become more economically viable.

Acknowledgements

Financial support from the College of Engineering and Physical Sciences and the Department of Chemical Engineering and Applied Chemistry, Aston University for MEng Research Project (D. J. Nouwe Edou) is gratefully acknowledged.
5. References


Declaration of interests
☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: